September 11, 2014

Response to M. Sasaki

We are grateful for this reviewer's comments on our manuscript. Based on their comments and suggestions, we have revised our manuscript in an effort to improve it and address their concerns. Below is our response to each of their comments (reproduced in bold).

1. Since diffusion rate in the air is much greater than that in water, the rate limiting process is diffusion of methane in water in air-water exchange. The diffusion rates of dissolved gases in water strongly depend on wind velocity. The effective thickness of diffusion layer in Eq.(1) in this manuscript might be change in wind velocity. Generally, since it is too difficult to measure the thickness of diffusion layer or to measure the concentration distributions of gases in the thin layer, the gas transfer coefficients k are empirically determined as proportional coefficients to the difference of bulk concentrations (chemical potentials) between air and water. Many values were proposed for CO2 exchange between air and ocean (Nightingale and Liss, 2003). I believe (Sasaki et al., 2010) that the air-lake transfer coefficient of CO2 proposed by Cole and Caraco (1998) is applicable to such small and shallow lakes in this manuscript, though the conversion of value for CO2 to that for CH4 is needed using Schmidt number. In this equation, the wave effect by wind (bubble formation with wave breaking) is evaluated weaker. I would like to ask authors to consider the wind effect on lake to air diffusive flux of methane in the future.

We appreciate the reviewer's suggestions and believe we have addressed their recommendation in our response to another reviewer's (V. Stepanenko's) comments (see Stepanenko Response p.10881, 1.12-16, also copied here below). We used the transfer coefficient from Cole and Caraco (1998) and converted it appropriately using the Schmidt number of CH<sub>4</sub>.

## Response to Stepanenko's similar comment:

Unfortunately, we did not measure wind speed at Goldstream Lake during the study period, so we are unable to apply a wind-dependent parameterization of the exchange coefficient in our model. Because Goldstream L. is surrounded by trees, we believe that the average wind speed at Goldstream Lake during the open-water periods is similar to that of the low-wind Mirror Lake, studied by Cole and Caraco (1998), so we revised our study to use the average value of the exchange coefficient they reported instead of the value of  $\delta_{eff}$  by Kling et al. (1992). This revision did not appreciably change our results, as the exchange coefficient calculated from the  $\delta_{eff}$  from Kling et al. differed by 2% from that from Cole and Caraco (1998).

We have included an additional section (A5) in our manuscript with a sensitivity analysis of how these approximations affect our results. As an extreme scenario, we assume that we grossly underestimated the magnitude of  $CH_4$  Diffusion emissions during the open-water periods and that all dissolved  $CH_4$  is released by diffusion during this time. In this case, the magnitude of open-water Diffusion emissions would double, and they would constitute 16% instead of 9% of

yearly  $CH_4$  emissions. We would like to emphasize that these approximations have no effect on our calculation of the magnitudes of IBS and Direct Ebullition emissions, which we consider to be the focus of this study.

2. There are no dissolved oxygen concentrations (DO) of water column during the icecovered season in this manuscript. When an oligotrophic lake is capped by ice cover, DO is supersaturated because of biological activity (perhaps, photosynthesis of mosses and algae at the bottom of lake) (Yoshida, et al., 1975). If similar phenomenon commonly occurs in seasonal ice-covered shallow lakes, the supersaturated DO must accelerate the methanotrophy of dissolved methane during the ice-covered season analyzed by Eq.(6) in this manuscript.

Our measurements (Fig. 8) indicate that the concentration of dissolved  $O_2$  is essentially zero during the majority of the ice-cover periods in this study. Snow covered the entire lake surface during this time, and our field observations indicate that essentially no sunlight penetrated the ice/snow layer, suggesting that photosynthesis did not occur during this time. Measured dissolved  $O_2$  concentrations begin to increase at the beginning of the ice-melt period in the spring, which we attribute to diffusion from the atmosphere through a degrading snow/ice layer.

We have added a sentence to our revised manuscript stating that measured dissolved  $O_2$  concentrations were similar to those calculated in the model (i.e., they were near-zero throughout most of the ice cover period).

We thank the reviewer for the time and thought they put into their comments, which have helped us improve our manuscript. We hope that our revised manuscript will be considered suitable for publication in *Biogeosciences*.